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AMORE Mo-99 Spike Test Results

Nuclear Engineering Division

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AMORE Mo-99 Spike Test Results

by
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January 2018

CONTENTS

1. INTRODUCTION **1**

2. METHODS **1**

3. RESULTS AND DISCUSSION **1**

3.1 LINAC 1
3.2 Gas Handling System 2
3.3 DU Target 3
3.4 Mo-99 Recovery Column 3

4. CONCLUSIONS **6**

REFERENCES **7**

FIGURES

1 Beam Power Delivered to the Target throughout the Irradiation Performed on October 19, 2017 2

2 Gas Concentration and LINAC Power as a Function of Time. 2

3 Mo-99 Stripping Curve for the First Titania Column. 4

TABLES

1 Result from the Mo-99 Recovery Column after the Irradiation on October 19, 2017. 3

2 Results from Mo-99 Spike Sampling Tests. 5

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1. INTRODUCTION

As a first step to produce Mo-99 in phase 2 of the Argonne Molybdenum Research Experiment (AMORE), we are performing a series of tests in which Mo-99 and natural Mo are added to a target vessel and the target solution is processed to recover Mo. This report describes a Mo-99 spike test with pH 1 sulfuric acid that was performed with an electron beam on a depleted uranium (DU) target. We ran previous spike tests without irradiating the sulfuric acid target solution [1]. We describe phase 2 of the AMORE system in our previous reports [1–4].

The Mo-99 recovery column was processed the day after the irradiation took place. This test was one of the final steps required to commission AMORE before adding uranyl sulfate solution to the phase-2 AMORE system. The Accelerator Readiness Review Committee will review results of the commissioning and overall readiness of the facility for normal operations. The Nuclear Engineering Division Director will send a letter documenting commissioning and results of the readiness review to our Chief Operations Officer. Once reviewed, a letter will be sent to the Argonne DOE Site Office informing them that the experiment has been commissioned, and the facility is ready for normal operation.

The total irradiation time was ~5 hours, and the maximum energy achieved was 20 kW, which was limited to ~10-minute increments due to heat dissipation limitations of the radiofrequency (RF) modulators. The composition of the headspace gas in the target vessel was monitored during irradiation, and the Mo-recovery system was operated the day after irradiation ended. The Mo-99 product was not processed through the concentration column due to an issue in the BigFoot hot cell. This has been remedied for future experiments.

2. METHODS

The system components (gas handling system, LINAC, DU target, separation system, and BigFoot hot cell) for phase 2 of the AMORE system were described in previous reports [1–4].

3. RESULTS AND DISCUSSION

3.1 LINAC

The beam energy for the irradiation performed on October 19, 2017 was 40 MeV. The macro pulse width was 5 μs, which corresponds to ~2.6 μCoulomb/pulse. The beam size at the target face was 24 mm by 21 mm. Figure 1 shows the beam power profile, where the total irradiation time was roughly 5 hours. The LINAC was stopped several times during irradiation (when the beam power is zero). Those stoppages happened because the DU target water temperature got too high (we set a very narrow operation range), and arcs occurred in an accelerating structure.

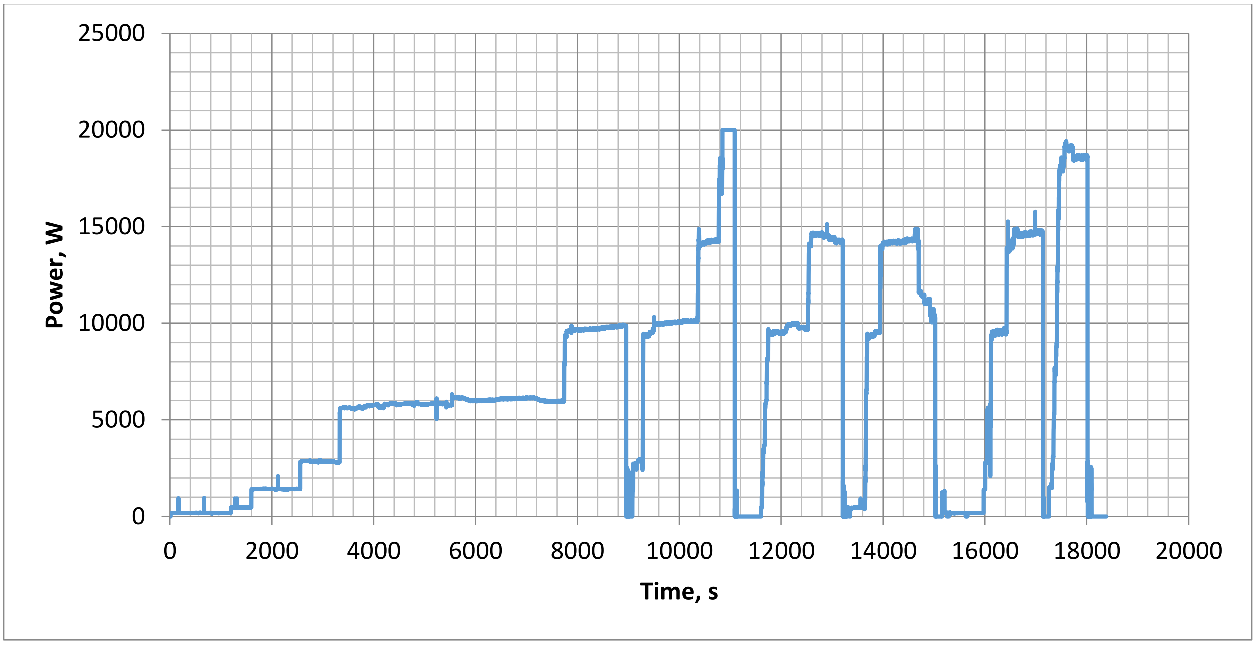

FIGURE 1. Beam Power Delivered to the Target throughout the Irradiation Performed on October 19, 2017.

3.2 Gas Handling System

Figure 2 shows the gas concentration relative to the LINAC power settings as a function of time. The experiment proceeded as expected; hydrogen build-up did not occur. The residual oxygen recombined with hydrogen, and no additional oxygen was added during irradiation. Hydrogen concentration was below the detection limit for the RGA (0.0063%) throughout the entire experiment.

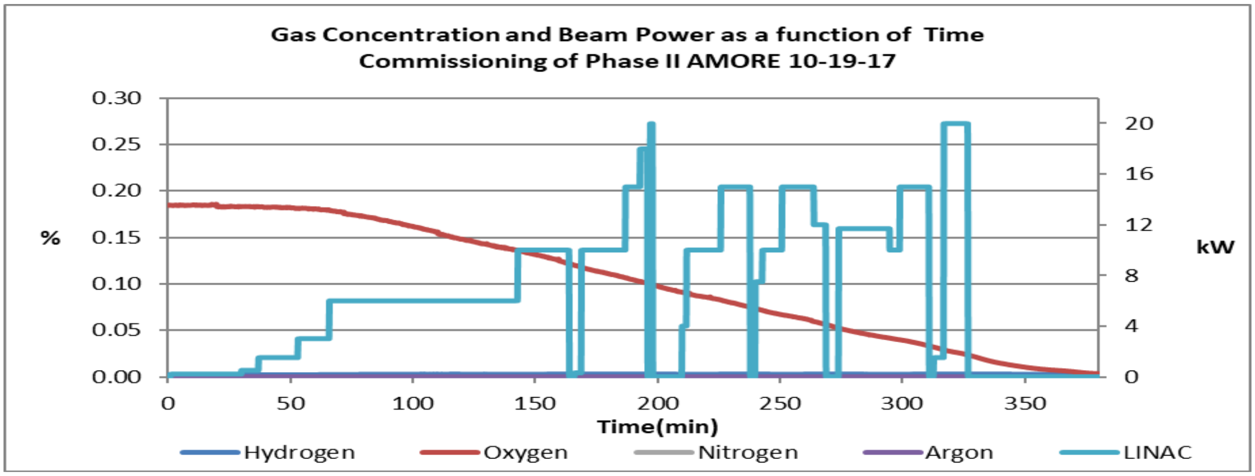

FIGURE 2. Gas Concentration and LINAC Power as a Function of Time.

3.3 DU Target

We verified performance of the DU target at several different power levels. The expected temperature difference between the inlet and outlet cooling water temperature was less than 1°C. During irradiation, the noise and uncertainty in the temperature measurements were larger than 1°C, so precise differential temperature measurements were not feasible. On a qualitative level, the difference in the inlet and outlet water temperature was on the order of 1°C.
After the irradiation and processing, we collected and analyzed a sample of the water from the target cooling loop with an HPGe. No radioactivity (from corrosion or fission products) was detected in cooling system.

3.4 Mo-99 Recovery Column

Table 1 provides the results for behavior of the recovery column. Activity in each stream was determined based on gamma counting of a small aliquot and total mass. The data in Table 1 show differing results for the feed to the Mo recovery column pre- and post-irradiation in the sampling loop. This is likely due to insufficient mixing and/or pulling residual Mo-99 spike solution into the system during the transfer from the verification tank to the target vessel, because this line was not properly washed. We also investigated whether the photon production of Mo-99 from the Mo-98 and Mo-100 found in the natural Mo added (3.5 mg) to target solution could affect the Mo-99 activity in the system. However, scaling production calculations from Phase I to Phase II predict that the total contribution from the $^{98}Mo(n,\gamma)^{99}Mo$ (the cross-section is 130 mb for thermal neutrons, but it also exhibits a 60-b distinct resonant peak at 12 eV and many

TABLE 1. Result from the Mo-99 Recovery Column after the Irradiation on October 19, 2017.

Sample	Mo-99 (mCi)	% Mo-99 based on pre-irradiation	% Mo-99 based on post-irradiation	% Mo-99 activity in each stream based on column effluents, column strip, column, and post-strip column effluent
Mo-99 Feed pre-irradiation	17.2			
Mo-99 Feed post-irradiation	20.6			
Mo-99 Feed Target Mixing Loops	15.1			
Column Effluent	0.20	1.16	0.97	1.4
Acid Wash	0.020	0.10	0.097	0.10
Water Wash 1	0.010	0.060	0.049	0.10
Strip—BigFoot Sample	14.3	83.1	69.4	96.8
Water Wash 2	0.009	0.052	0.044	0.10
Base Rinse	0.16	0.93	0.78	1.10
Sorbent	0.07	0.40	0.30	0.50

more resonant peaks in the range of 400 eV to 40 keV, some of them above 100 b) and $^{100}Mo(\gamma,n)^{99}Mo$ (peak cross-section of 142 mb at 14 MeV) reactions only accounts for a combined production of 1.5–3 μCi [5,6]. Therefore, the small quantity of Mo-99 produced would not affect the post-irradiation sample to an appreciable degree. The difference in pre- and post-irradiation Mo-99 values is another indication of sampling and/or mass balance problems in the system. The spike test confirms our previous experiments showing that due to an unavoidable residual volume in the sampling loop, the error in the activity of samples taken by this method is 10–20% low.

Based on the average of the pre- and post-irradiation feed samples, the Mo-99 recovery from the first titania column was unaccountably low; it was only 76%, which is significantly less than that achieved in phase-1 experiments and other phase-2 Mo-99 spike tests. However, as shown in Table 1, very little Mo-99 was present in any of the other effluent streams or the sorbent itself. Based on the sum of all effluent streams and what remained on the column following processing, the Mo-99 yield in the strip was 97%, which is a typical yield.

Based on the results of previous spike tests and ancillary experiments, we reduced the stripping volume from 20 column volumes (CVs) to 10 CVs for this experiment. The stripping curve shown in Figure 3 suggests that all of the Mo-99 was recovered in the first 10 minutes, which is equivalent to about 7 CVs of 1 M NaOH.

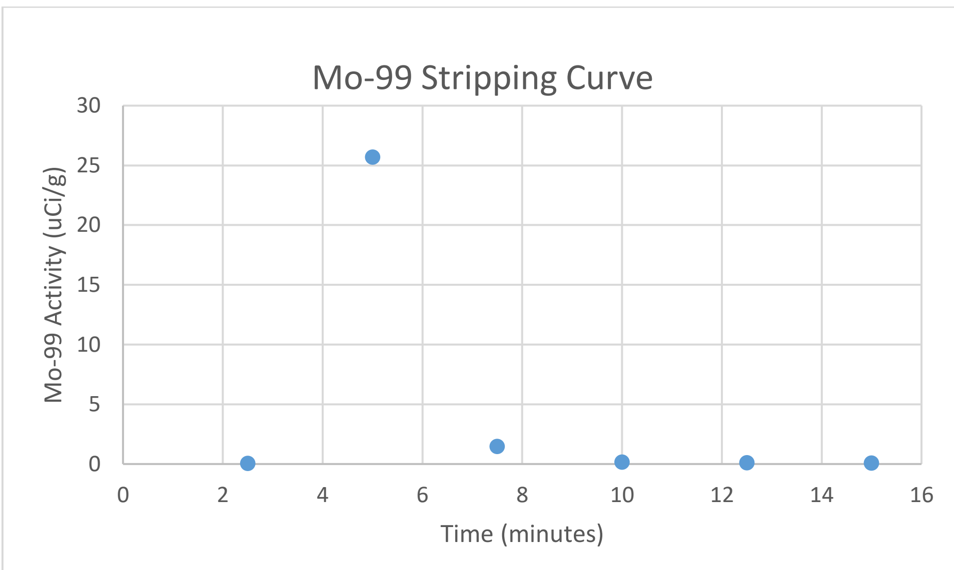

FIGURE 3. Mo-99 Stripping Curve for the First Titania Column.

To better understand the discrepancy in Mo-99 activity for the different feed samples collected during the Mo-99 spike test with irradiation and processing, we performed a second test without processing and irradiation. In this test, we added an additional Mo-99 spike to the pH 1 sulfuric acid solution, and several samples were collected. In the spike test with irradiation and processing, there was a difference of nearly 20% between the sample collected from the verification tank and the sample collected from the target vessel after processing. Table 2 shows the Mo-99 activities for samples taken via the syringe port after mixing in the verification tank for 2 hours at 300 mL/min, samples being transferred from the verification tank to the target vessel, and samples being transferred from the target vessel to the verification tank. We also collected a second set of samples that were identical to the first—except mixing in the verification tank only lasted 1 hour at 300 mL/min. The activity was in the first sample collected is about 12% higher than the average activity obtained in the last four samples collected, and is the highest overall. The Mo-99 activity in the second sample was also high compared to the activity in the majority of the samples. Along with results from the first sample, this may indicate insufficient mixing. The last four samples collected give us confidence that a representative sample can be obtained during transfer from the target vessel to the verification tank via the syringe port if sufficient mixing (at least 4 hours at 300 mL/min) has taken place. Based on this, we recommend that in future uranyl-sulfate irradiations, the post-irradiation feed sample should be collected during transfer from the target vessel to the verification tank via the syringe port. This will cause prevent ~100–200 mL of feed solution from passing through the Mo recovery column.

TABLE 2. Results from Mo-99 Spike Sampling Tests.

Sample	Decay Corrected Activity (mCi/g)	Mass of Solution (g)	Total Activity (mCi)
2 hours of mixing in verification tank	2.72	17165	46.7
Solution transferred from verification tank to target vessel	2.65	17165	45.5
Solution transferred from target vessel to verification tank	2.41	17135	41.3
1 hour of mixing in verification tank	2.41	17135	41.3
Solution transferred from verification tank to target vessel	2.43	17135	41.6
Solution transferred from target vessel to verification tank	2.49	17100	42.6
Average Mo-99 Activity in last four samples			41.7

4. CONCLUSIONS

We achieved the maximum power deposition of a 20-kW beam on the DU target for 10 minutes during the ~5-hour irradiation. The LINAC performed well, and it achieved the expected beam profile. The DU target cooling water was sampled post-irradiation, and no gamma activity was found, which suggests that integrity of the target was not compromised. The gas analysis and collection systems performed well, the catalytic recombiner effectively recombined the hydrogen and oxygen, and there was no large buildup of hydrogen or oxygen during the irradiation.

Based on the data obtained from the two spike tests performed, we learned several lessons:

- Sufficient mixing (at least 4 hours at 300 mL/min) is necessary to collect a representative sample of the feed.

- The spike test confirms our previous experiments. The results of this test were obtained using a dye that, due to an inconsistency in residual volume in the sampling loop, results in an error of 10–20% in the activity of samples taken using this method.

- When adding a Mo-99 spike into the system using a syringe, the line needs to be washed with 3–5 syringe volumes to guarantee proper introduction of the spike into the system.

We determined that after 4 hours of mixing, a representative sample of feed solution can be obtained during transfer of the solution from the target vessel to the verification tank. The next AMORE experiment will be with uranyl sulfate in the target vessel and is expected to take place in February 2018.

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